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ON THE APPLICATION OF CHEMISTRY TO ARCHAEOLOGY

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INTRODUCTION

Archaeology may be defined as the science which yields information and develops theories about past human activity by means of a study of ancient material remains. As a general rule the archaeologist is much more interested in the form or style of such remains than in their chemical composition, for this may often have little or no archaeological significance. For example, the form of flint implements may be an important index to the cultural status of a certain group of early people, but it is improbable that any significant information from the standpoint of archaeology could be obtained from an investigation of the chemical composition of the flint of such implements. But sometimes important archaeological information may be obtained by a study of the chemical composition of material remains. This information may supplement that obtained from a study of form or style, or it may be entirely new information not otherwise obtainable. The purpose of the present paper is to indicate by means of general categories, specific illustrative examples, and a select bibliography how chemical facts, methods, and theories have been and may continue to be applied to the study of the material remains of past human activity for the purpose of furthering archaeological science. Also indicated are miscellaneous ways in which chemistry may be useful to the field and to the museum archaeologist. For the most part the illustrative examples are based upon the writer's own experience, though some mention is made of pertinent work of other investigators.

THE IDENTIFICATION OF MATERIALS

Perhaps the simplest and most obvious application of chemistry to archaeology is the exact qualitative identification of materials in order that these may be accurately described in archaeological publications. The accurate identification of materials is important also because significant interpretations may depend upon the exact nature of certain materials. Errors of identification, some of them rather glaring from the chemical viewpoint, are not infrequent in the older archaeological literature, and are by no means absent from some of the recent literature. Copper is sometimes confused with bronze, bronze with brass, lime mortar with gypsum plaster, solid bitumens with vegetable resins, and so on for other materials superficially similar. Such confusions have arisen either because no proper identification of a material was ever made in the first place or because the archaeologist, even after a correct identification had been made, did not then appreciate the importance of the chemical distinction between certain materials very similar in appearance and put to similar uses. Actually, the inexact or loose identification of the kinds of materials found in excavations may lead to serious error. For example, it might not seem important to determine whether a given object is composed of brass or of bronze, but a very wrong conclusion as to time of manufacture could be reached by a failure to determine which it is, or a failure to appreciate the significance of the distinction, brass having been first made about the middle of the first century B. C. and bronze some twenty centuries earlier.

Though a few materials, such as wood, may be readily recognized, and even identified as to variety, by means of physical appearance alone, it is unsafe as a general rule to rely solely on physical properties for the identification of ancient materials. Even when the material of an object may be recognized by simple inspection, the application of chemical tests may be very desirable both to confirm and to amplify the observation. For example, there is generally no difficulty in recognizing an ancient iron object as such even when it is in the usual highly oxidized condition, but for iron objects of early date it is important to determine whether they were fashioned from meteoric iron or from manufactured iron, the difference in cultural level represented by the use of the one kind as contrasted to the other being very great.

The problem of making a correct identification of ancient materials found in excavations may be very different from the problem of identifying the same materials in a fresh condition. Thus, for example, a clean piece of copper may be differentiated from a clean piece of bronze at a glance because of the difference in color, but no such difference is apparent when the two materials are in a highly oxidized condition, as they usually are when found in excavations. Furthermore, in the identification of materials found in excavations it is not so important to determine what a material is now, as it is to determine what it was originally before it underwent a series of chemical changes. During the excavations at the site of the ancient Agora in Athens, Greece, an object was found in the form of a thin shield. This was described as a bronze shield before a chemical examination was made. Actually no free metal of any kind could be found in it and no compounds of tin could be found among the copper corrosion products. The conclusion was that it was originally a shield of beaten copper. Though the original nature of inorganic materials may usually be determined with little difficulty, it is otherwise with organic materials for these are not only more likely to undergo extensive chemical changes but such changes are generally much more complicated.

For materials that are deceptively similar to each other in appearance chemical tests are of course a necessity for the purpose of differentiation. Silver containing a moderate proportion of alloy cannot with certainty be distinguished from fine silver on the basis of appearance, though such a distinction may be very significant when the objects are in the form of coins. Still more necessary is identification by chemical tests for those materials the appearance of which gives little or no clue as to their actual nature. For example, a white material in the form of lumps and fine powder was found in ornamental ceramic containers in graves of Greek women buried about the fourth century B. C. in Attica. Also found were various other articles of the toilet. After some speculation as to whether the white material was chalk or white clay, chemical analysis showed that it was artificial basic lead carbonate or white lead, evidently used as cosmetic powder.

It is important to note that the exact chemical identification of ancient materials may lead to various inferences and interpretations that are fully as important, or even more important, than the mere establishment of identity for the purpose of correct description. Thus the preceding example of the identification of a material as white lead immediately implies the discovery and use of a process for its manufacture, its sale as an article of commerce, and the employment of a poisonous substance as a cosmetic that may well have had an adverse effect on the public health.

INDICATIONS AS TO SOURCES OF MATERIALS, THE EXISTENCE OF COMMERCE IN PARTICULAR MATERIALS, AND THE DIRECTION OF TRADE ROUTES

The presence of numerous specimens of a material of foreign origin at an archaeological site is a clear indication of the import of such a material through trade or commerce. If the source of this material can also be located then the direction or path of this trade or commerce can be established also. Sometimes the chemical examination of materials can determine whether they are of foreign origin or can supplement other evidence indicating a foreign origin.

Deposits of native copper do not occur in Ohio, yet a large number of artifacts of native copper have been found at various sites in the state. The only possible explanation is that native copper must have been brought into the state, either by trade or in this particular case by expeditions to the source. That the deposits of the Lake Superior region were the source of this copper receives additional confirmation from the fact that the chemical composition of the copper of the artifacts is the same as that of the copper of the Lake Superior deposits.

At the Agora excavations in Athens a number of specimens of a bright red pigment were found which on analysis turned out to be cinnabar, the native sulfide of mercury. The character of the geological formations in Greece do not indicate that cinnabar ever occurred there in appreciable quantities, and therefore it was concluded that in all probability this was an imported pigment. The discovery of ancient cinnabar workings is Asia Minor supports this conclusion and in addition indicates the probable existence of an export trade in cinnabar from Asia Minor to Athens.

True Baltic amber is rather easily differentiated from other fossil resins because it yields a considerable proportion of succinic acid on dry distillation. Objects of fossil resin, identified by this characteristic as Baltic amber, have been found in considerable numbers at prehistoric sites in Greece. In spite of the considerable distance involved, either by the sea route or by travel across the then undeveloped land areas of Europe, the conclusion is that a commerce in amber must have existed between the Baltic coast and the Mediterranean region even in these remote prehistoric times. Indication of the existence of a trade in the reverse direction, at least at a much later period, is given by the composition of small brass objects dating from the opening centuries of the Christian Era which have been found in the eastern Baltic coastal region. There is no evidence that such brass was of native manufacture, and indeed it is improbable that it was in view of the cultural state of the inhabitants of the region at that period. The significant fact is that the brass of these objects was found to have the same composition as the brass of the Roman coins of the same period. From this it has been reasonably concluded that Roman coins reached this distant region by trade and were converted into these objects by the inhabitants.

EXPLANATION OF UNUSUAL PHENOMENA ENCOUNTERED IN THE COURSE OF EXCAVATION

Slow chemical changes extending over long periods of time not infrequently result in unusual phenomena which may lead to erroneous conclusions if not interpreted correctly. For example, an ancient weight found during the excavation of the Agora in Athens appeared to consist of bronze or copper, but a chemical examination showed that it was actually a lead weight coated superficially with a thin plating of metallic copper and with corrosion products of copper. The lead itself was of the usual high degree of purity. From a consideration of the facts it seemed most likely that the thin plating of copper was formed by electrolytic displacement when copper bearing solutions came into contact with the lead object, and that the copper corrosion products were formed by the subsequent corrosion of part of the electrolytically deposited copper. The copper bearing solutions were probably produced through contact of ground water with neighboring bronze or copper objects. In view of the fact that the layer of copper and copper corrosion products on the lead object was still very superficial in spite of burial for approximately 2000 years, it is very probable that the copper bearing solutions were extremely dilute and that as a result the rate of the plating process was exceedingly slow. Similarly, what appear to be bronze or copper examples of well known ancient silver coins have been found which on examination have proved to be actually such silver coins superficially coated with metallic copper and its corrosion products. This type of plating apparently occurred because such coins were simultaneously in contact with copper bearing solutions and an active metal.

Sometimes well-crystallized minerals are found in association with ancient objects, and it may wrongly be concluded that these minerals were originally associated with the objects. The presence of such minerals is more often the result of their formation by natural means near, on, or even in ancient objects after their burial. However, the objects themselves often play a necessary part in the formation of the minerals. For example, the formation of cuprite or azurite on or near bronze objects depends primarily of the material of these objects. Occasionally the association of certain substances with objects occurs because of some very unusual or special condition. For example, a dark brown compound was found in the layers of corrosion products on lead objects from one part of the Agora excavation area. This brown compound was identified as lead dioxide. In view of the fact that this area was adjacent to, and in part actually included, a section of the road bed of an electric railway line it seemed probable that stray electric currents were the essential agency in the formation of the lead dioxide on the lead objects.

DETERMINATION OF THE USES OF OBJECTS

Though the uses to which ancient objects were put is often a matter readily decided by simple inspection, a chemical examination may sometimes be helpful in establishing the exact use to which a given object or a given kind of object was put. This is especially true for containers of various sorts. For example, in the excavations at the Athenian Agora numerous fragments of empty jars were found on the interior surfaces of which was a black or dark brown resinous coating. Since the unglazed pottery body of such jars was porous it was evident that the purpose of the internal resinous coating was to render them impervious to liquid. Impressed stamps on the handles of the jars showed that they were of foreign origin, and it therefore was further evident that they were containers in which some liquid product was imported. Obviously they were not, originally at least, containers used for water. Experiments on the solubility of the resinous coating in various liquids showed that it was more or less readily soluble in vegetable oils. Hence the vessels could not have been used as containers for olive oil or any other vegetable oil. Though soluble to some extent in concentrated alcoholic solutions, the resinous coating was not appreciably soluble in dilute alcoholic solutions containing less then 20% by volume. Therefore it appeared very likely that these jars were containers in which wine was imported. The resinous coating was found to consist of fused mastic resin.

As another example may be cited the results of an examination of the contents of a small jar found in the excavations at Corinth. This was a small round jar of green-glazed pottery with a very narrow neck. It was of Byzantine manufacture and probably of the 12th century A. D. Such vessels have long been classified as pomade jars, but no concrete evidence as to what they originally contained had ever been produced. When found this particular jar appeared to be intact since it was obviously full of something, and its neck was tightly sealed with what appeared to be a corroded iron stopper. However, on opening the vessel it was seen that the rusted mass which filled the neck was simply a part of the remains of an original iron applicator rod that had stood in the vessel and had fitted loosely in the neck. This rod had expanded on rusting and consequently had filled the opening. The vessel itself was found to be filled with fine clay which evidently had infiltrated into the empty or nearly empty jar before the neck had become sealed through rusting and expansion of the iron rod. The hope of finding the jar full of its original contents or the modified remains of these contents therefore met with disappointment. However, the clay from the vessel was extracted with boiling ether and on evaporation of the ether extract there was obtained a considerable quantity of a fatty material that was evidently the remains of the original material left in the vessel before it became filled with clay. This fatty matter was found to consist largely of a mixture of fatty acids which from their properties were judged to be the result of the hydrolysis of the glycerides of a vegetable oil or a mixture of vegetable oils. Evidence obtained by chemical means therefore gave a firm basis to the previous opinion that vessels of this type were used as containers for oils or oily preparations such as might be used for dressing the hair.

DETERMINATION OF THE DETAILS OF ANCIENT TECHNICAL PROCESSES

The scientific examination of ancient objects can yield information about the technique of their manufacture that cannot otherwise be obtained. For example, the microscopic examination of metal objects, coins for example, by the methods of metallography can show whether they were produced by casting or by striking, or by a combination of casting and striking. Likewise the examination of weapons yields interesting details on the method of manufacture. It has been observed, for example, that the edges of bronze or copper weapons were often subjected to severe cold work such as might have been done by long continued hammering, whereas the body of the weapons largely retained their original cast structure. Evidently the hardening effect of cold working was well recognized and deliberately utilized.

Sometimes modern experiments in imitation of ancient processes yield information on the details of the procedures that must have been employed in order to achieve the results now seen in ancient objects. On ceramic bodies and glazes a considerable number of such experiments have been made. It has been found, for example, that the black glaze on certain types of ancient pottery could only have been produced by firing in a reducing atmosphere.

DETERMINATION OF CHRONOLOGICAL SEQUENCES

Chemical methods of investigation are apparently not of general use for the determination of the chronological sequences of ancient objects, though for certain special kinds of objects they may be of considerable use, at least in connection with other kinds of evidence. For example, it was found as a result of quantitative chemical analyses of a large number of ancient Greek bronze coins from different localities that there exists a broad general relationship between the date of production of such coins and their chemical composition. Coins of the earliest date generally contain the highest proportion of tin and very little or no lead. Somewhat later coins contain a lower proportion of tin and a moderate proporation of lead. The latest coins contain the lowest proportion of tin and a very high proportion of lead. Furthermore, a rather sharp change in the composition of these coins was observed to have taken place shortly after the middle of the second century B. C. which may be ascribed to the fall of Carthage and the consequent stoppage in the shipment of tin by the sea route from Britain. This relationship between the chemical composition of the coinage alloy and its date of production has been successfully applied, in connection with other evidence, to the arrangement in chronological sequence of a considerable number of types of the late bronze coins of Athens.

Chemical methods of investigation are sometimes useful for fixing the earliest possible date at which a given object could have been manufactured. For example, it appears to be definitely established on the basis of analyses of numerous dated objects that zinc was not introduced into alloys produced in Mediterranean counries until after the middle of the first century B. C., and therefore the presence of zinc as an essential component in the metal of an undated object from any of these countries is an almost certain indication that this object was not made before this time.

INDICATIONS OF CULTURAL LEVEL AND ECONOMIC CONDITION

The presence or absence of certain kinds of materials may be an index of the cultural status of a people. Obviously the absence of metal among the remains of a given people indicates a low state of technical development and a low state of general culture. The greater the variety of metals and alloys utilized by a people the greater their technical development, and, as a rule, the higher their general cultural development. The same appears to be true for certain kinds of non-metallic materials such as glazes and glasses. Some materials that serve as indices of this nature may, of course, be recognized without chemical aid, though the application of chemical methods may make their recognition more certain. On the other hand, some kinds of materials that are indicative of cultural level are not easily recognized for what they are without chemical aid. For example, the introduction of lead or tin into glazes, or the introduction of certain of the less common elements into glasses, represent technical advances which may be, and in fact sometimes have been, overlooked in the study of ancient remains. An increase in the variety of materials utilized by a people in the course of their existence may be taken as an indication of a rise in cultural development and conversely a decrease may be taken as an indication of a decline.

It is probable that a rise or fall in the variety of materials utilized is also in general an indication of a corresponding rise or fall on economic status, but a more direct indication of this may be obtained through a chemical study of the particular kind of objects that bear a direct relationship to economic status, namely, the coins of a people. For example, the fineness of Roman silver coins underwent a slow but almost steady decrease that paralleled the increasing economic difficulties that occurred with the decline of the empire until finally nothing but silver-coated coins came into use. Sudden changes in the composition of coins may mark sudden economic difficulties or disaster. As illustrative of this an ancient example seems almost inadequate in view of the many striking examples afforded by the numismatic history of Europe in recent years.

EXPLANATION OF THE ALTERATIONS IN THE APPEARANCE OF ANTIQUITIES AS CONDITIONED BY ENVIRONMENT AND TIME

During long burial in the ground a great variety of chemical reactions occur between the material of most objects and the surrounding soil and ground water. In order fully to explain the appearance of objects found in excavations, to deduce with confidence their probable original appearance, or to decide upon the proper method of restoration, it is often essential to understand the nature and course of these chemical reactions. The appearance and general condition of objects of the same material and of the same age taken from different excavation sites may be very different. Thus a simple tin bronze from one site may be coated with a hard coherent layer of corrosion products of the sort commonly called a patina, whereas a similar bronze of the same age from another site may be coated with loose porous masses of corrosion products. The very extent to which objects of a given material undergo corrosion at one site as compared to those at another site may vary enormously. For example, archaeologists observed that bronze found at Corinth was almost invariably in a severely corroded state, whereas similar bronze of equal or greater age found at certain other sites in Greece was not so corroded. A special investigation into this phenomenon showed that the ground water at Corinth happens to contain an abnormally high concentration of chloride, and that this is sufficient to account for the severe corrosion of the buried bronze.

Chemical considerations may also serve to correct common misconceptions as to the original appearance or nature of certain materials or objects. By reason of its usual beautiful iridescent appearance ancient glass, for example, is regarded by some as a product artistically superior to modern glass, and one that cannot be successfully imitated by modern glass makers. Observation has shown, however, that ordinary modern soft glass also becomes iridescent when buried in certain soils, especially those slightly alkaline because of the presence of ammonia from decaying organic matter. Experiments have also shown that this action may be greatly hastened by treatment with alkaline solutions at an elevated temperature. All such action consists in the removal of the metallic ingredients from the surface of the glass leaving thin films of silica on the surface which refract light in spectral colors. If ancient iridescent glass has any real artistic superiority over modern glass made iridescent by chemical treatment it is because the ancient glass has been subjected by natural conditions to a very slow and long continued action which has caused a deeper and perhaps somewhat different type of leaching of the surface.

RESTORATION AND PRESERVATION OF ANTIQUITIES

In general, the deterioration of buried objects is predominantly the result of chemical change, and it is therefore reasonable to expect that the restoration of such objects should be best effected by chemical treatment. The deterioration of bronze or copper, for example, is primarily the result of oxidation, and the reverse process of reduction should tend to restore this to its original condition. This principle has been applied extensively to the restoration of bronze and copper objects. In practice this is done by electrolytic procedures, in one of the best of which the corroded object is made the cathode, an inert electrode of nickel or platinum is used, and the electrolyte is a dilute solution of sodium hydroxide. On passage of a feeble current for a long time, often several weeks, the corrosion products on the object are slowly reduced back to metal, much of which plates back on the object in approximately its original position. At the same time any clay or sand entangled in the corrosion products falls to the bottom of the electrolytic bath. When reduction is complete the object is scrubbed to remove nonadherent matter, thoroughly rinsed with distilled water, and dried in an oven. The results achieved by this means are much superior to those obtained by former methods of mechanical cleaning.

Even when objects are of such nature that they undergo no appreciable chemical deterioration during burial, they may become coated with foreign matter that requires chemical treatment for its safe and effective removal. For example, once there was found an important inscribed Sumerian cone of baked clay that was extensively incrusted with a thick and adherent deposit of calcium carbonate and calcium sulfate which could not be removed mechanically without flaking off the softer inscribed surface of the cone. Treatment with hydrochloric acid readily removed the deposit without damaging the cone. Similarly, incrustations of calcium carbonate on pottery are now often removed on a large scale at excavation sites by treatment with hydrochloric acid.

Whether ancient objects are restored by chemical means or not, problems concerning their preservation for purposes of exhibition or study often arise, and these are frequently best solved by the application of chemical principles. In ordinary air the corrosion of an ancient metal object, for example, may continue at an appreciable rate unless proper precautions are taken. For such an object this problem is sometimes best solved by coating it with a protective layer of wax or synthetic resin. Where such treatment is not desirable or feasible the object may be kept in a sealed glass case containing an inert gas, or in a case containing a powerful desiccating agent that keeps the air very dry. Sometimes special problems arise such as the so-called bronze disease, or cylic corrosion, of bronze or copper objects. Such a problem can only be solved by application of chemical principles.

Occasionally certain problems of this sort not directly connected with ancient objects, yet of interest to the archaeologist, may also be solved by chemical means. The preservation of paper impressions of inscriptions on stone is an example. These paper impressions, which are commonly used in the study of lapidary inscriptions, are obtained by applying one or more large sheets of chemical filter paper over the inscription, thoroughly wetting the paper, pressing it into the carved letters on the stone, letting it dry in place, and then peeling off the dried paper. In this way an exact reverse impression of the inscription is taken. Unfortunately, these convenient copies are fragile and there is a tendency for fine details to be obliterated on frequent handling, or even on storage under humid weather conditions when a number of impressions are piled together. It was found that by spraying such impressions with a special methyl methacrylate lacquer they could be stiffened and toughened to such an extent that these disadvantages connected with their use were completely eliminated.

DETECTION OF FALSE ANTIQUITIES

The forgery or alteration of ancient objects presents a problem that plagues the dealer in antiquities, the museum archaeologist, and sometimes even the field archaeologist. Frequently a counterfeit ancient object may be detected by chemical tests or analyses. As an example may be cited the means by which an alleged ancient Greek bronze statuette, offered for sale to a museum director, was shown to be not as represented. Though satisfactory from the standpoint of style, this object appeared suspicious because of the thinness of the corrosion products or patina on its surface. On analysis of a small sample of the metal taken from an inconspicuous place in the base it was found that this metal lacked the usual impurities such as arsenic, gold, nickel, and silver commonly present in small proportion in genuine ancient bronze. Furthermore, the alloy was found to contain zinc as a major component which failed to agree by at least several centuries with its alleged date. On these bases it was concluded that it very probably was an object of modern manufacture.

Certain physico-chemical methods of examination, either alone or in connection with chemical tests, are useful for detecting forged antiquities and especially for detecting modern additions, alterations, or repairs. Examination under ultraviolet light, for example, is especially useful for the critical examination of ivories and marbles. Under ultraviolet light different marbles fluoresce different colors, and the surface of ancient marble frequently has a characteristic fluorescent color which is very different from that of fresh stone of the same kind. On this basis alterations or repairs in an ancient marble object which are not at all visible in ordinary light may become strikingly conspicuous when the object is viewed under ultraviolet light. Similarly, infrared light and X-rays have been found useful in the detection of falsification.

CONCLUDING REMARKS

The above general categories and the illustrative examples, of which many more could be given, sufficiently show that chemistry may be of important service to archaeology. The titles in the appended bibliography, which is by no means exhaustive, further show the aid that chemistry may render to archaeology. As far as the writer has been able to determine this is the first general bibliography on this special field of investigation. A peculiarity of the published work in this field is its scattered nature and the consequent difficulty of locating and assembling it. Much has been published in obscure periodical works and in the form of pamphlets in small editions. Another considerable proportion has been published in the form of extensive footnotes or appendices in archaeological works. Because of the nature of the subject and the way in which much of it has been published a good many items have escaped the usual scientific abstract journals and general indices. As indicated by the extent of this bibliography much work has already been done in the past. Much more remains to be done, however, and it should be possible eventually to erect a considerable body of special knowledge having its own data, techniques, and general rules. For such a body of special knowledge, a distinct and separate branch of applied chemistry in itself, an appropriate descriptive name would be *archaeological chemistry.*

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